PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 2005M015	**FOR FURTHER ACTION** See Form PCT/IPEA/416	
International application No. PCT/EP2005/000948	International filing date *(day/month/year)* 28.01.2005	Priority date *(day/month/year)* 22.03.2004
International Patent Classification (IPC) or national classification and IPC INV. C07C45/50 C10G3/00 C07C45/00		
Applicant EXXONMOBIL CHEMICAL PATENTS INC.		

1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 6 sheets, including this cover sheet.
3. This report is also accompanied by ANNEXES, comprising:
   a. ☒ *sent to the applicant and to the International Bureau)* a total of 3 sheets, as follows:
      ☒ sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).
      ☐ sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.
   b. ☐ *(sent to the International Bureau only)* a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in electronic form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).

4. This report contains indications relating to the following items:

☒	Box No. I	Basis of the report
☐	Box No. II	Priority
☐	Box No. III	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
☐	Box No. IV	Lack of unity of invention
☒	Box No. V	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
☐	Box No. VI	Certain documents cited
☐	Box No. VII	Certain defects in the international application
☐	Box No. VIII	Certain observations on the international application

Date of submission of the demand	Date of completion of this report
20.01.2006	04.07.2006
Name and mailing address of the international preliminary examining authority: European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Authorized officer Bertin-van Bommel, S Telephone No. +31 70 340-4231

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/EP2005/000948

Box No. I Basis of the report

1. With regard to the **language**, this report is based on

   ☒ the international application in the language in which it was filed

   ☐ a translation of the international application into , which is the language of a translation furnished for the purposes of:
   - ☐ international search (under Rules 12.3(a) and 23.1(b))
   - ☐ publication of the international application (under Rule 12.4(a))
   - ☐ international preliminary examination (under Rules 55.2(a) and/or 55.3(a))

2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report)*:

   Description, Pages

   1-23 as originally filed

   Claims, Numbers

   1-17 received on 26.01.2006 with letter of 20.01.2006

   Drawings, Sheets

   1/1 as originally filed

   ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing

3. ☒ The amendments have resulted in the cancellation of:
   - ☐ the description, pages
   - ☒ the claims, Nos. 17,18
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
   - ☐ the description, pages
   - ☐ the claims, Nos.
   - ☐ the drawings, sheets/figs
   - ☐ the sequence listing *(specify)*:
   - ☐ any table(s) related to sequence listing *(specify)*:

* *If item 4 applies, some or all of these sheets may be marked "superseded."*

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1-17
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-17
Industrial applicability (IA)	Yes:	Claims	1-17
	No:	Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Re Item V

Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents:
D1: US-A-4 593 127 (BUNNING ET AL) 3 June 1986 (1986-06-03)

Novelty

1. With its example 1, figure 2 and table 2, the document D1 discloses a continuous process for the hydroformylation of propylene comprising feeding a stream containing 98% propylene (78) and a first (76) and second (82) synthesis gas stream with a H_2/CO ratio of 1.08 to two serially connected hydroformylation reactors (D1: primary and secondary systems), both comprising a Rh catalyst.

2. The subject-matter of claim 1 firstly differs over that of D1 in that the feed rate of the propylene stream equals at least 3 tonnes per hour, i.e., the process of claim 1 is carried out at industrial scale (description: page 3, ln. 17-20). The process of D1 is one of bench scale.

3. Secondly, the subject-matter of claim 1 differs over that of D1 in condition (b) stating that the molar ratio $(H_2+CO)/C_3H_6$ ratio contained in the **fresh feed to the *process*** is greater than 1.93 and at most 2.00. In D1, table 2, said ratio equals 1.89 for the primary system and 2.01 for the combined primary and secondary system.

4. Claim 1 is therefore new in view of document D1 (Article 33(2) PCT).

Inventive Step

5. The wording of claim 1 does not exclude a subsequent secondary reactor as described in D1 to be incorporated in the process of claim 1. In fact, the description of the present application (p.13, ln.12-22) teaches the use of more than one reactor in series as a

preferred embodiment.

Because the ratio of condition (b) in claim 1 refers to the feed to the *process*, rather than the reactor, the combined primary and secondary systems of D1 is considered in the assessment of the inventive step of claim 1 in view of D1.

6. As mentioned above, the difference of the subject-matter of claim 1 over D1 is the feed rate of 3 tonnes per hour, i.e. industrial scale vs bench scale of D1 and the feed ratio of syngas over propylene of at most 2.00 vs 2.01 in D1.

7. The problem to be solved by the present application is considered to be the achievement of a high propylene conversion on an industrial scale.

8. The person skilled in the art of hydroformylation would, in order to scale up the process of D1 to industrial scale, as according to customary practice, increase the feed rate to at least 3 tonnes per hour, without the exercise of inventive skill.

As to the remaining difference of the ratio, there is no evidence of a technical effect of using a ratio of at most 2.00 compared to using a 2.01 ratio as disclosed in D1. Moreover, as can be expected from the different scales, the propylene conversion of the combined primary and secondary system in D1 is higher than the conversions reached in the present application (see table 1 on page 16 of present application, compared to below calculation based on table 2 of D1):

Conversion in D1:
Ingoing stream of propylene (IN): 76 ; 1.764 gmole/hr
Outgoing streams of propylene (OUT): 67,89,91,92 ; total of 0.026833 gmole /hr

Conversion = (IN-OUT)/IN = 98.48%, compared to 94.56% in the present application.

9. Therefore, because there is no unexpected effect to the conversion that could be attributed to selecting the syngas:propylene feed ratio up to at most 2.00, and because this slight difference in feed ratio is considered to fall within acceptable variations that are to be expected when scaling up a process without any surprising effect, no inventive step can be

recognised for the subject-matter of claim 1 (Article 33(3) PCT.

10. Dependent claims 2-17 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step, as they are either disclosed in D1 (see the corresponding passages cited in ISR), or because they are part of customary practice followed by the person skilled in the art.

Miscellaneous

11. It is noted that, assuming the basis exists in the application as filed, as required by 19(2) PCT, were the wording of claim 1 have been unambiguous about the conditions (a), (b) and (c) relating to the hydroformylation *reactor* per se, rather than the conditions relating to the *process*, an inventive step would probably be recognised, since the subject-matter of claim 1 would be compared to only the primary system of D1, rather than the entire *process* comprising also the secondary system. This would result in a different comparison of feed ratio's and conversions, showing an unexpected effect. However, as presently worded, the conditions relate to the entire process and the subject-matter of claim 1 is compared to the disclosure of D1, accordingly.

CLAIMS

1. A continuous process for the hydroformylation of propylene comprising feeding

(i) a propylene stream at a rate of at least 3 tonnes per hour, and

(ii) synthesis gas comprising hydrogen and carbon monoxide

to a hydroformylation reactor in which the propylene is hydroformylated over a rhodium containing catalyst, characterised in that

(a) the propylene stream contains at least 97 mole % of propylene;

(b) the molar ratio of (H_2+CO) contained in the fresh synthesis gas fed to the process, to the propylene contained in the fresh propylene stream fed to the process, is greater than 1.93 and at most 2.00; and

(c) in the synthesis gas feed, the hydrogen over carbon monoxide molar ratio is from >1:1 to 1.2:1

2. The process according to claim 1 in which the propylene stream contains at least 99.5 mole % of propylene.

3. The process according to claim 1 or 2 in which the sulphur content of the propylene feed is not greater than 500 ppb by weight, calculated on an atomic basis.

4. The process according to any of the preceding claims in which the reactive nitrogen content of the propylene feed is not greater than 10 ppm by weight, calculated on an atomic basis.

5. The process according to any of the preceding claims in which the chlorine content of the propylene feed is not greater than 5 ppm by weight, calculated on an atomic basis.

6. The process according to claim 3, 4 or 5 wherein the propylene feed contains, by weight and on an atomic basis, less than 50 ppb sulphur or less than 50 ppb reactive nitrogen or less than 50 ppb chlorine.

7. The process according to claim 6 wherein the aggregate weight content in the propylene feed of sulphur and reactive nitrogen and chlorine, on an atomic basis, is less than 50 ppb.

8. The process according to any of the preceding claims in which the hydroformylation catalyst is an oil-soluble rhodium complex comprising a low valence rhodium (Rh) complexed with a triorganophosphorus compound.

9. The process according to claim 8 in which the triorganophosphorus compound comprises an oil-soluble triarylphosphine, trialkylphosphine, alkyl-diaryl-phosphine, aryl-dialkylphosphine, triorganophosphite or bisphosphite containing, per molecule, one or more phosphorus atoms capable of complexing with Rh.

10. The process according to claim 9 in which the triorganophosphorus compound is triphenylphosphine or 6,6'-[[3,3',5,5'-tetrakis (1,1-dimethylethyl)-1,1'-biphenyl-2,2'-diyl] bis (oxy)] bis-dibenzo [d,f] [1,3,2]-dioxaphosphepin.

11. The process according to any of the preceding claims in which the Rh concentration in the hydrofomylation reaction mixture is in the range of from 1×10^{-5} to 1×10^{-2} moles/litre.

12. The process according to any of the preceding claims in which the hydroformylation is carried out at a temperature in the range of from 40 to 200°C.

13. The process according to any of the preceding claims in which the hydroformylation is carried out at a pressure in the range of from 0.05 to 10 MPaa.

14. The process according to any of the preceding claims in which the carbon monoxide partial pressure in the reactor is not greater than 50% of the total pressure.

15. The process according to any of the preceding claims wherein the propylene feed stream contains up to 5000 ppb by weight of dimethyl ether.

16. The process according to any of the preceding claims wherein the molar ratio of (H_2+CO) to propylene is greater than 1.94.

17. The process according to any of the preceding claims wherein the molar ratio of hydrogen to carbon monoxide is from 1.1:1 to 1.2:1.